Optical studies of MOVPE grown GaN layers*

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Photoluminescence and reflectance studies of MOVPE grown GaN samples were performed. From reflectance measurements optical constants were calculated by means of Kramers-Krönig analysis in the energy region 0–6 eV.

Keywords: GaN, MOVPE, photoluminescence, reflectance, Kramers-Krönig analysis

1. Introduction

GaN and related compounds currently attract extensive attention because of their potential device application. The wavelength of radiation from GaN-based group III nitride semiconductors can be tuned over the wide range from visible to ultraviolet by alloying and forming heterostructures with other nitrides (AlN and InN). Because of that, these materials are very attractive for short-wavelength optical applications such as blue-UV light emitting diodes, blue-UV laser diodes [1,2], video displays, full colour TV system etc.

In this paper we report photoluminescence and reflectance investigations of GaN samples made by MOVPE process in which the additional buffer layers were inserted between the low temperature buffer layer and the high temperature GaN overlayer grown on it. From reflectance spectra measured in wide spectral region (2–6 eV) optical constants (the refractive index n and the extinction coefficient k) were calculated. Photoacoustical investigations of GaN sample were also made.

2. Experimental

2.1. Samples growth

GaN layers were grown in the atmospheric pressure, single wafer, and vertical flow MOVPE system. The low frequency (20 kHz) inductive heating method was used to raise the temperature of graphite susceptor up to 1050°C. GaN epitaxial films growth was performed with H2 carrier gas. Trimethylgallium (TMGa) and ammonia (NH3) were used as gallium and nitrogen sources. Typical flow rates were in the ranges of 1-2 l/min for the carrier gas, 0.5-6 l/min for ammonia, and 1-7 ml/min for hydrogen bubbling, through TMGa kept at t = -14°C. The films were deposited on differently oriented sapphire substrates under identical process conditions to allow the comparison of the crystal quality and to find correlation with the process parameters. However, the optimisation of the growth procedure was made taking as criteria only GaN layers parameters grown on c sapphire substrate. Several parameters were changed to optimise the GaN growth process. Just before the growth process, the substrates were degreased in organic solvents (trichloroethane, acetone, isopropyl alcohol), etched in a hot (180°C) solution of H₂SO₄:H₃PO₄ (3:1) rinsed in de-ionised water, and dried in the filtered nitrogen. Comparing

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to the conventional two-step (with one buffer layer) growth procedure [3], the second buffer layer, grown during the temperature raise before annealing of GaN buffer was added. At the same time the V/III ratio was linearly changed from 8300 at the beginning of step 4 to the infinity (the TMGa flow was almost vanishing) at its end. The only purpose of this stage is to prevent the thermal desorption and mass transport from the optimised very thin (~20 nm) low temperature buffer layer before the subsequent high temperature GaN overlayer is grown. We observed that this multi--buffer layer strategy influenced mainly structural, electrical, and also optical properties [4,5] of the high temperature GaN layers. As was obtained from X-ray diffraction measurements, adding the second buffer layer allowed us to grow layers with reduced mosaicity and smaller value of c-lattice parameter variations compare to those grown without the additional buffer [4].

2.2. Photoluminescence measurements

The photoluminescence (PL) measurements were held at the room temperature. As an excitation beam the 351 nm Ar⁺-ion laser line was used. All samples were excited with the same power of the excitation beam. PL signal was analysed by 2-m double grated monochromator with Hammamatsu photomultiplier as a detector.

2.3. Reflectance measurements

The reflectance measurements were held at the room temperature in the energy region 2–6 eV in nonpolarizeed light at the near-normal incidence. The reflectance signal from the investigated sample was compared, at each measurement point, to the reflectance from aluminium plate to obtain the absolute value of reflectance. The monochromator slits were open wide enough to average interferences from the sample.

3. Results and discussion

Near-band-edge UV emission line is visible at the energy 3.41 eV in PL spectra taken at room temperature (RT) from all investigated samples. It is difficult to say if the dominant transition in undoped GaN at RT is band-to-band or excitonic transition [6]. The energy position of this line agrees with the value of GaN energy gap equal to 3.41 eV and obtained by Powell et al. [7] and is in disagreement with the value

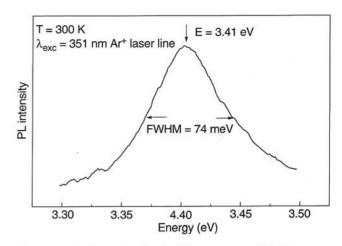


Fig. 1. Typical near-band-edge PL spectrum of GaN at room temperature.

of 3.42 eV obtained by Monemar et al. [8] for GaN on sapphire. The differences in these values can be explained by the presence of the strain in the layers. The FWHM of this line changes from 74 meV to 95 meV from sample to sample (depending on growing conditions). Such values (especially lower of them) indicate good quality of investigated samples. Typical PL spectrum at RT is presented in Fig. 1.

In Fig. 2 the reflectance spectrum of GaN sample is presented. One can see the exciton structure at the energy gap. Besides this structure the spectrum is rather flat, slowly increasing with the energy increase. Up to 6 eV no other structure is expected because transitions at the edges of the Brillouine zone should be observed in the vacuum ultraviolet.

In order to calculate the optical constants by means of Kramers-Kröning (KK) analysis the measured spectrum was extrapolated below the lowest energy point

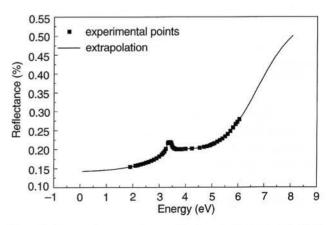


Fig. 2. Room temperature reflectance spectra of GaN sample. Squares indicate measurement points and the solid line is the fit to the measured spectrum and extrapolation below and above it.

and above the highest one [9,10]. The determined extrapolations are also presented in the Fig. 2. It should be undertaken that in this case the measured reflectance increased at the high energy end. Therefore the high energy extrapolation must be able to continue this growth. Only the non-typical solution, presented in paper [10], enable us to provide good results in such a situation. The determined n and k values are presented in Fig.3. The value of n increased from n = 2.2 to $n \approx 3$ with a small structure at the Γ point. The value of k is equal to zero below the energy gap (it was an assumption used in KK analysis).

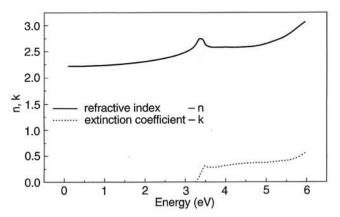


Fig. 3. Optical constants of GaN sample calculated from the reflectance spectrum by means of Kramers-Krönig analysis.

4. Conclusions

GaN layers have been grown by MOCVD using new multi-buffer layer strategy. Photoluminescence investigations show good quality of the obtained samples. Both in photoluminescence and reflectance spectra a feature near to bandgap energy of GaN can be observed. Kramers-Krönig analysis was used to calculate the refractive index n and the extinction coefficient k in the energy region 0–6 eV.

References

S. Nakamura, T. Mukai, and M. Senoh, "Candela-class high brightness InGaN/AlGaN double heterostructure blue-light emitting diodes", Appl. Phys. Lett. 64, 1687–1689 (1994).

- S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Tamada, T. Matsushita, H. Kiyoku, and Y. Sugimoto, "InGaN-based multi-quantum-well structure laser diodes", *Jpn. J. Appl. Phys.* 35, L74 (1996).
- 3. O. Briot, J.P. Alexis, S. Sanchez, B. Gil and R.L. Aulombard, "Influence of the V/III molar ratio on the structural and electronic properties of MOVPE grown GaN", *Solid State Electr.* 41, 315–317 (1997).
- 4. J. Kozłowski, R. Paszkiewicz, R. Korbutowicz, M. Panek, B. Paszkiewicz, and M. Tłaczała, "Structural properties of MOVPE GaN layers grown by a new multi-buffer approach", MRS Internet J. Nitride Semicond. Res. 3, 27 (1998).
- M. Ciorga, L. Bryja, J. Misiewicz, R. Paszkiewicz, R. Korbutowicz, M. Panek, B. Paszkiewicz, and M. Tłaczała, "The influence of MOCVD process scheme on the optical properties of GaN layers", will be published in *Mat. Sc. Eng. B*.
- M. Smith, J.Y. Lin, H.X. Jiang, and M. Asif Khan, "Room temperature intrinsic optical transition in GaN epilayers: The band-to-band versus excitonic transitions", *Appl. Phys. Lett.* 71, 635–637 (1997).
- 7. R.C. Powell, N.E. Lee, Y.W. Kim, and J.E. Greene, "Heteroepitaxial wurtzite and zinc-blende structure GaN grown by reactive-ion molecular-beam epitaxy: Growth kinetics, microstructure, and properties" *J. Appl. Phys.* 73, 189 (1993).
- B. Monemar, J.P. Bergaman, T. Lundstrom, C.I. Harris, H. Amano, I Akasaki, T. Detchprohm, K. Hiramatsu, and N. Sawaki, "Optical characterisation of GaN and related materials", *Solid State Electr.* 41, 181–184 (1997).
- 9. K. Jezierski, "A linear-equations algorithm for reflectivity extrapolation determination in Kramers-Krönig analysis", *J. Phys. C: Solid State Phys.* 17, 475–482 (1984).
- 10. K. Jezierski, "Improvements in the Kramers-Krönig analysis of reflection spectra", *J. Phys. C: Solid State Phys.* **19**, 2103–2112 (1986).